Magnetic extrema in electronic susceptibility and heat capacity of mesoscopic systems

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Abstract

Oscillating behaviour of the susceptibility χ and heat capacity C is considered for normal and superconducting mesoscopic systems (nanoclusters and quantum dots). It is proved that at low temperature an increasing magnetic field applied to a mesoscopic system generates local extrema of χ and C. A *maximum* for χ and a *minimum* for C simultaneously arise in those points of the field where crossings of quantum levels of the normal and superconducting mesoscopic systems take place.

Key words: Nanoclusters, quantum dots, magnetic oscillations, level crossings
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1 Introduction

Low temperature oscillations of diverse electromagnetic and thermal characteristics (resistivity, magnetization, susceptibility, thermal conduction, heat capacity etc.) of macroscopic and mesoscopic bodies in an increasing uniform magnetic field are apparently a general property of systems formed by charged particles. These oscillations have been last years studied experimentally and theoretically in mesoscopic systems of different geometry and microscopical structure: in mesoscopic rings [1], in a thin spherical layer [2], in superconducting discs [3,4,5,6,7], in micron-size heterostrucures [8]. Observing such variety of properties and objects one is forced to arrive at a conclusion that there must be a common reason for arising this phenomenon. In this Letter as a source of such oscillations we regard crossings of the quantum levels of a mesoscopic

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system abstracting from the physical nature of the levels and system itself. We concentrate here on static properties (the magnetic susceptibility χ and heat capacity C) in the regime of the thermodynamical equilibrium of the canonical ensemble at low temperatures.

We assume that a system exposed to a uniform magnetic field (B) possesses a number of discrete quantum states altering their mutual energy disposition depending on B. Thereby for each value of B there exist a lowest state, the ground state for a given B, and the nearest to it first excited state with energies $E_0(B)$ and $E_1(B)$ respectively. Each of these energies is a smooth function of B, however $E_0(B)$ and $E_1(B)$ can locally have opposite slopes with B. Therefore the difference $E_{10}(B)$

$$E_{10}(B) = E_1(B) - E_0(B); \quad E_{10}(B_0) = 0. \tag{1}$$

can become equal to zero for a value of $B = B_0$.

In Sec.2 we show that this point of the level crossing is a peculiar point where at very small temperatures both χ and C reach local extrema: a *maximum* for χ and a *minimum* for C.

2 Extrema in χ and C

In the low temperature limit only two first terms practically exhaust the canonical partition function

$$Z = exp[-\beta E_0(B)] + exp[-\beta E_1(B)] + Z_{\mathrm{r}}, \quad \beta = 1/T. \tag{2}$$

The rest, Z_{r}, gives a weak background for χ and C near the point B_0 in the vicinity of which the partition function can be expressed via a small parameter ξ:

$$\begin{aligned} \xi &= \frac{1}{2}\{exp[-\beta E_{10}(B)] - 1\}; \quad \xi(B_0) = 0, \\ \ln Z &= -\beta E_0(B) + \ln(1+\xi) + \alpha + \ln 2, \\ \alpha &= \frac{1}{2}\sum_{i\geq 2} exp\{-\beta(E_i - E_0)\} \ll 1. \end{aligned} \tag{3}$$

The magnetic susceptibility and heat capacity are naturally divided near B_0 into smoothly varying parts χ_{s}, C_{s} and the rapidly varying functions χ_{e}, C_{e} with extrema in B_0:

$$\chi(B_0) = \chi_{\rm s} + \chi_{\rm e}, \tag{4}$$

$$\chi_{\rm s} = -\frac{1}{V}\frac{\partial^2(E_0 - \alpha T)}{\partial B_0^2}, \quad \chi_{\rm e} = \frac{1}{\beta V}\frac{\partial^2 \ln(1+\xi)}{\partial B_0^2}, \tag{5}$$

$$C(B_0) = \quad C_{\rm s} + \quad C_{\rm e}, \tag{6}$$

$$C_{\rm s} = \beta^2\frac{\partial^2\alpha}{\partial\beta^2}, \qquad C_{\rm e} = \beta^2\frac{\partial^2 \ln(1+\xi)}{\partial\beta^2}. \tag{7}$$

V is the volume of the system. The derivative $\partial/\partial B_0$ implies hereafter the derivative with respect to B in the point of $B = B_0$.

At low temperatures ($T < \delta$, δ is the mean level spacing at $B = 0$) we take into account only those terms in Eqs.(5) and (7) which are proportional to the maximum power of β at $B = B_0$, $\partial^n\xi/\partial B_0^n \simeq 0.5(-\beta)^n(\partial E_{10}/\partial B_0)^n$, that gives

$$\chi_{\rm e}(B_0) = \frac{\beta}{4V}\left(\frac{\partial E_{10}}{\partial B_0}\right)^2 > 0, \tag{8}$$

$$\frac{\partial\chi_{\rm e}}{\partial B_0} = 0, \quad \frac{\partial^2\chi_{\rm e}}{\partial B_0^2} = -\frac{\beta^3}{8V}\left(\frac{\partial E_{10}}{\partial B_0}\right)^4 < 0, \tag{9}$$

$$C_{\rm e}(B_0) = 0, \tag{10}$$

$$\frac{\partial C_{\rm e}}{\partial B_0} = 0, \quad \frac{\partial^2 C_{\rm e}}{\partial B_0} = \beta^2\left(\frac{\partial E_{10}}{\partial B_0}\right)^2 > 0. \tag{11}$$

$\chi_{\rm e}$ is paramagnetic ($\chi_{\rm s}$ determines the diamagnetic component of χ) and proportional to the squared difference of the magnetic moments of the crossing states. Thus, Eqs.(8) -(11) confirm the existence of a local maximum in χ and a minimum in C at the level crossing point B_0. Increasing the field can generate appearance of new level crossing points that will give χ and C an oscillating functions of B. Intervals between peaks and their amplitudes depends on the physical nature of the system and its shape. Here we limit ourselves by considering weak fields (the cyclotron radius is much larger than the size of the system). The alterations of quantum levels v.s. B are caused by removing the spin and rotational degeneracy, the latter occurs when a system has rotational symmetry. To investigate oscillating behavior of χ and C in Sec.3 we apply the independent electron model to normal mesoscopic systems and in Sec.4 superconducting clusters of the perfect spherical shape are described by using the exact solution of the conventional superconducting hamiltonian.

3 Magnetic oscillations in normal clusters

The character of oscillations in electron χ and C of a mesoscopic system is dependent on the its shape. In spherical clusters at $B = 0$ each many–fold degenerate level (the spin-orbital interaction is omitted here) displays a sheaf-wise splitting. Hence magnetic sublevels originated from one spherical l-shell (l is an orbital momentum) cannot cross each other with the growth of B. Thus the minimum strength of a magnetic field, $B_{\min}$, (here we consider the case of the closed Fermi–shell at $B = 0$) required to arouse the first crossing and the first extremum is determined by $\delta_{\rm sh}$, the distance between the Fermi–shell, l_F–shell, and the unoccupied l_{F+1}–shell at $B = 0$: $\mu_B^* B_{\min}(l_F + l_{F+1} + 2) = \delta_{\rm sh}$, $\mu_B^* = \mu_B m^*/m$, m^* is the effective electron mass. We assume that orbital and spin projections are aligned at the first crossing. Nevertheless the absolute value of $B_{\min}$ is practically independent of l, since the level splitting near F at $B = 0$ is proportional to l: $\delta_{\rm sh} \simeq (2l_F + 1)\delta$, where $\delta = 4\varepsilon_F/3N$ is the mean level spacing of the Fermi gas near the Fermi energy (ε_F). Thus, $B_{\min} \sim \delta m^*/\mu_B m$ and for nanometer normal metallic clusters ($N \simeq 10^5$, $\varepsilon_F \sim 10\ eV$ and $m = m^*$) $B_{\min}$ is of the order of $1T$. The evolution of some single–electron levels in a spherical cavity and corresponding extrema in χ and C are shown in Fig.1.

Properties of quantum dots are frequently interpreted in the two-dimensional cylindrically symmetric oscillator model [9]. This symmetry also gives a rather high degeneracy of electron levels near the Fermi level, which is removed in the same manner as in the previous spherical case. $B_{\min}$ of the first level crossing is assessed analogously: $B_{\min} \sim \varepsilon_F m^*/2N\mu_B m$, N is the free electron number in the quantum dot. As ε_F and m^*/m for such systems can be small (e.g. for GaAs $m^*/m \sim 0.07$, $\varepsilon_F \sim 10\ meV$ [9]) oscillations of χ could be observable in relatively weak fields for a wide range of N. χ and C v.s. the cyclotron frequency are shown in Fig.2,3 for $N \simeq 10^3$ (we consider the case of the closed cylindrical Fermi–shell at $T = B = 0$). The feature of χ and C oscillations in systems with cylindrical or spherical symmetry is the presence of several groups of peaks corresponding firstly crossings of levels from two nearest cylindrical or spherical shells separated at $B = 0$ by one shell gap, then cross levels separated at $B = 0$ by two shell gaps and so on. Increasing temperature (Fig.3) leads to confluence of separate peaks so that curves for χ and C become similar and their gross structure is caused by the shell structure of the single–particle energy spectrum.

Much simpler behavior of χ and C as function of B is observed in clusters having no rotation symmetry axis. In this case level crossings are caused by the Zeeman splitting and perturbation of orbital motion by weak fields proves to be insignificant, because orbital energy shifts for such systems are quadratic in B. Then signs of energy denominators appearing in perturbative calculations

are opposite for levels near the Fermi–level that reduces second order terms. The example of such "spin" oscillations of χ is concerned with electron system in the oscillator well with oscillator frequencies $\Omega_x = 0.9\Omega_y = 0.5\Omega_z$ that corresponds to an ellipsoid with semiaxes $a_x \simeq 1.1a_y = 2a_z$ (we adopt the relation between semiaxes and oscillator frequencies from Ref. [10]: $\Omega_x a_x = \Omega_y a_y = \Omega_z a_z$). Such system has no rotation axis and a magnetic field directed along x–axis generates "spin" oscillations (Fig.4). These oscillations of χ in the anysotropic oscillator are much like oscillations in the equal level spacing model [11].

4 Magnetic oscillations in the gapless superconducting region

Another example of magnetic oscillations of χ and C is given by the single-shell model (SSM) of superconductivity. Such model can be a reasonable approach to a description of spherical superconducting clusters with numbers of conduction electrons $10^3 < N < 10^5$ [12]. There is a wide range of orbital momentum l_F for states successively becoming the Fermi level with growth of N. Averaging through N gives $\widetilde{l}_{\rm sh} \simeq N^{1/3}$ [12]. At $B = 0$ the level spacing between degenerate shells $\delta_{\rm sh} \simeq 2\widetilde{l}_F\delta \simeq 8\varepsilon_F/3N^{2/3}$ is much more than the BCS superconducting gap at $B = T = 0$, $\Delta_{\rm BCS}(0)$, e.g. $\delta_{\rm sh} > 10$ meV and $\Delta_{\rm BCS}(0) < 1$ meV for Al-clusters with N under consideration. Therefore the superconducting pair correlations can arise between electrons (their number is $N_{\rm sh}$) occupying only one shell with a large l.

The Hamiltonian of SSM consists of the pairing interaction with the strength G and the interaction with a field B. The exact spectrum of such Hamiltonian is well known [10] and at $B = 0$ is determined by the seniority ν (unpaired electron number)

$$E_\nu = -\frac{G}{4}(N_{\rm sh} - \nu)(4l + 4 - N_{\rm sh} - \nu). \tag{12}$$

For each ν there is a multiplet of states (degenerate at $B = 0$) with different orbital and spin projections which are splitted by a field. The energy of the lowest state for each ν is

$$E_\nu(\omega) = E_\nu - \frac{\omega}{4}\{\nu(4l + 2 - \nu) - \frac{1}{2}[1 - (-)^\nu] + 4(1 - \delta_{\nu.0})\}, \tag{13}$$

Thus with growth of B (in this Section $\omega \equiv \hbar\omega\mu_B^* B$) the role of the ground state is successively played by states with $\nu = 0, 2, 4, \ldots$ for even N_{sh} or $\nu = 1, 3, 5\ldots$ for odd $N_{\rm sh}$, i.e. increasing B leads to a sequence of crossings

of the ν-level with $\nu+2$-level, resulting in a step–wise decrease of the pairing gap (the region of the gapless superconductivity).

To take into account the temperature dependence of χ and C quantities we have analytically summed up those components of the canonical partition function Z which involve states inside the considered l–shell

$$Z = \sum_{\nu} exp(-\beta E_\nu)[\Phi_\nu - \Phi_{\nu-2}(1-\delta_{\nu.0})], \qquad (14)$$

$$\Phi_\nu = \sum_{\sigma_{\min}}^{\nu/2} \frac{2}{1+\delta_{\sigma.0}} \widetilde{\Phi}_{\nu/2+\sigma} \widetilde{\Phi}_{\nu/2-\sigma} \cosh(2\beta\omega\sigma),$$

$$\sigma_{min} = \frac{1}{4}[1-(-)^\nu],$$

$$\widetilde{\Phi}_k = \delta_{k.0} + (1-\delta_{k.0}) \prod_{\mu=1}^{k} \frac{\sinh(\beta\omega\frac{2l+2-\mu}{2})}{\sinh(\beta\omega\frac{\mu}{2})}, \quad \beta = \frac{1}{k_B T}.$$

Fig.5 shows χ and C as functions of the magnetic field and temperature. The latter is measured in $T_{\rm crit}$, the critical temperature of the conventional BCS theory for half–filled shell. Oscillations of χ and C created by level crossings in SSM spread on a range of rather small fields $\omega \simeq \Delta_{\rm BCS}(0)/\tilde{l}_F$ [12] ($B < 0.5\ T$ for Al-clusters with $N > 10^3$ and $m = m^*$). In nonspherical superconducting clusters where a weak magnetic field does not influence orbital motion, the level crossings are determined only by alignment of spins. Therefore greater values of $B_{\rm min}$ are expected to induce the first extrema in χ and C.

5 Conclusion

In conclusion we have shown that an increasing uniform magnetic field applied to a mesoscopic system transforms the structure of the ground state in a such a way that in level crossing points synchronously appear local extrema in magnetic susceptibility (*maxima*) and heat capacity (*minima*). Thereby these thermodynamic quantities oscillate with growth field. Periodicity and amplitudes of the low temperature oscillations carry information concerning properties of the quantum states of investigated normal and superconducting systems.

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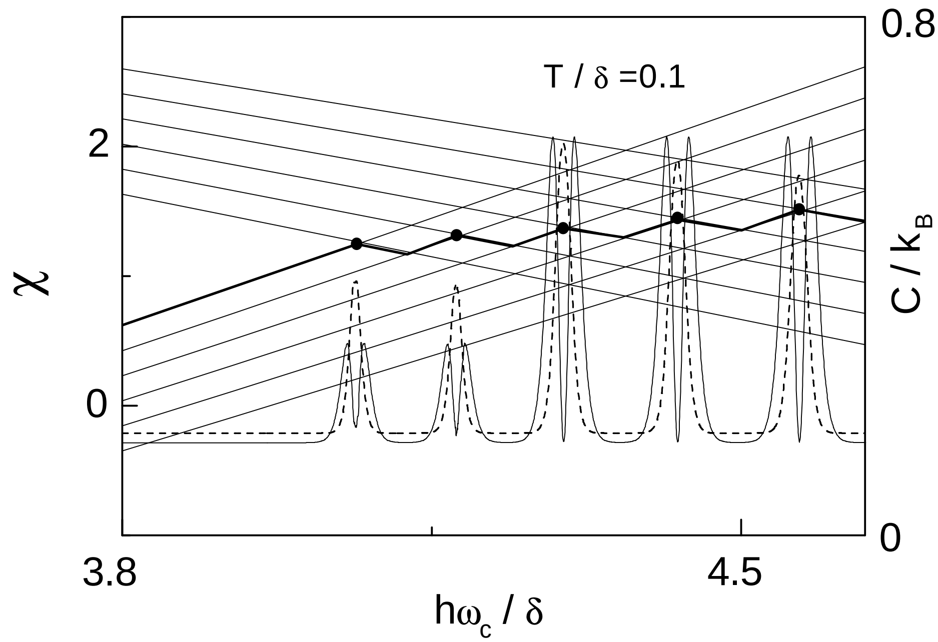

Fig. 1. Electron levels vs the cyclotron frequency ($\hbar\omega_c = 2\mu_B^* B$, $\mu_B^* = \mu_B m^*/m$) and magnetic oscillations of the susceptibility χ (dotted line) and heat capacity C (solid line) for a sphere with $N \simeq 10^5$. Points of level crossings where χ and C reach local extrema are marked by solid circles. $\delta = 4\varepsilon_F/3N$. The bold line corresponds to the Fermi-level. χ is given in units $10^4\cdot \mid \chi_{\rm L} \mid$, $\chi_L = -\mu_B^2 N/2V\varepsilon_F$ is the Landau diamagnetic susceptibility of the degenerate free electron gas, $m = m^*$.

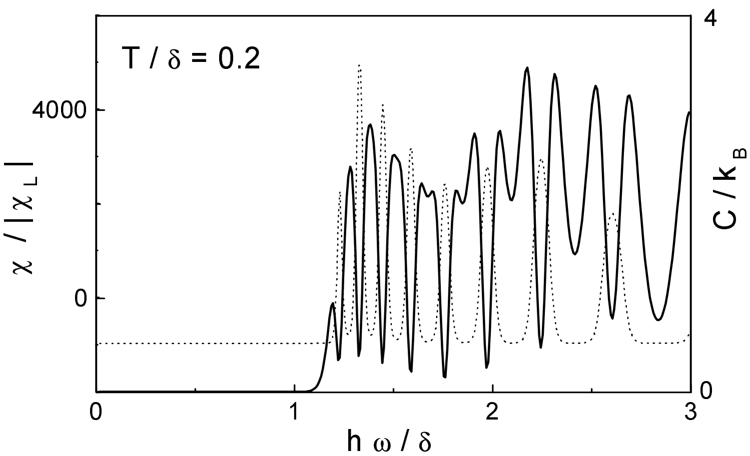

Fig. 2. Low temperature magnetic oscillations χ (dotted line) and C (bold line) in a two-dimensional cylindrical oscillator with $N \simeq 10^3$. $\delta = 4\varepsilon_F/3N$. χ is given in χ_{L}.

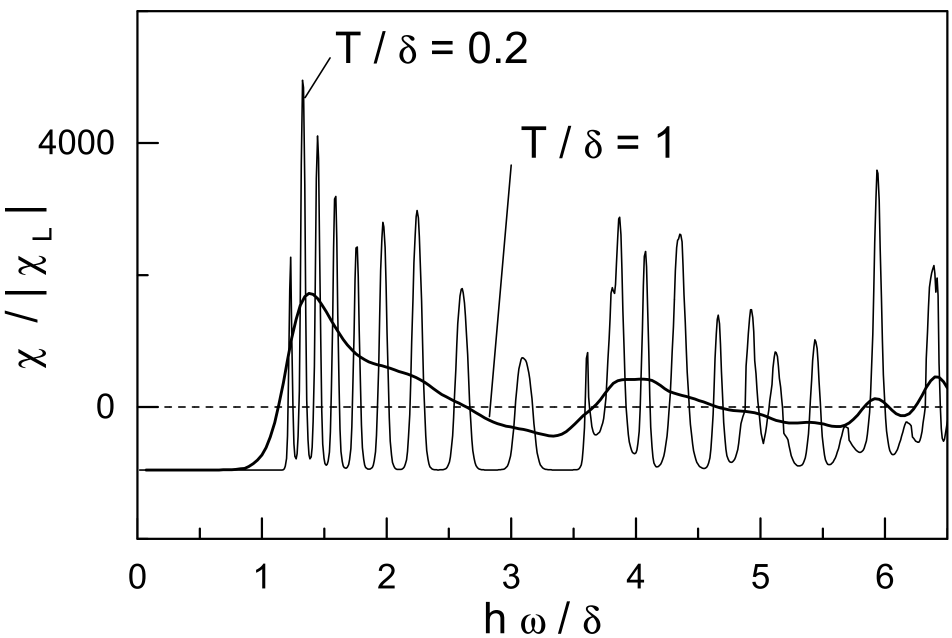

Fig. 3. χ vs the cyclotron frequency ω_c at different temperatures for a two-dimensional cylindrical oscillator with $N \simeq 10^3$ (closed Fermi-shell) $\delta = 4\varepsilon_F/3N$.

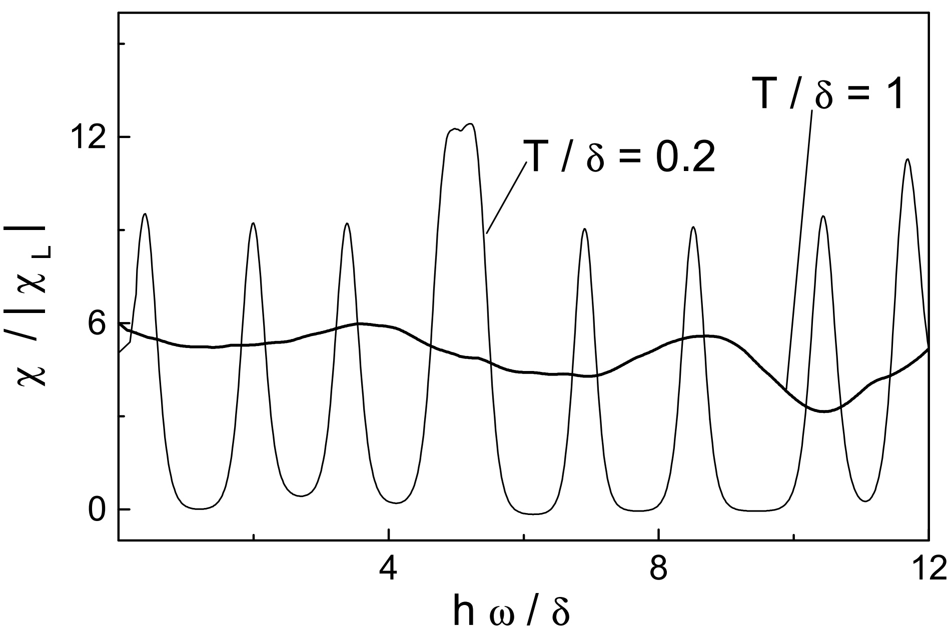

Fig. 4. χ vs ω_c at different temperatures for an anysotropic oscillator $(\Omega_x = 0.9\Omega_y = 0.5\Omega_z)$ with $N \simeq 10^5$, magnetic field is directed along x -axis.

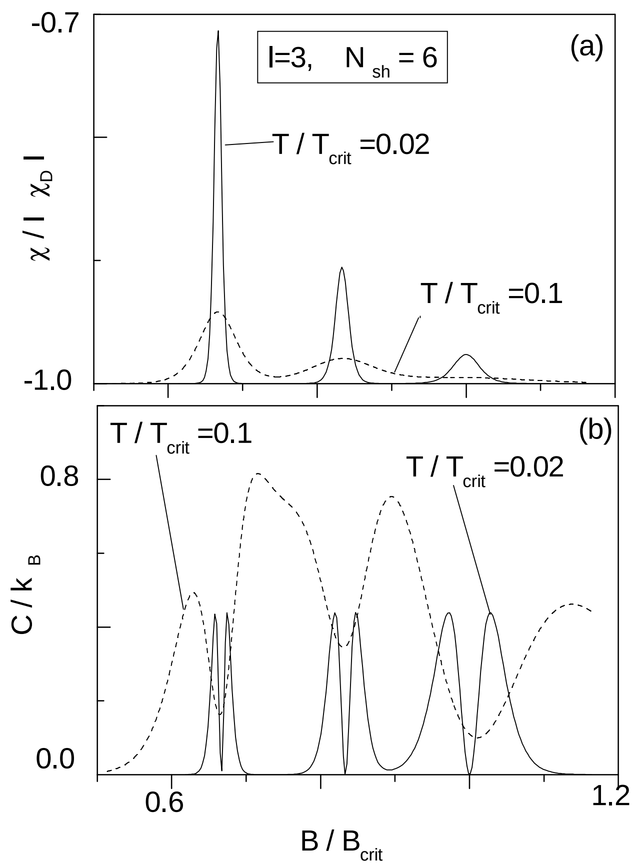

Fig. 5. Magnetic oscillations in a superconducting sphere. $B_{\rm crit}$ is the critical magnetic field of the BCS theory at $T=0$, $\Delta_{\rm BCS}(T=0,B_{\rm crit})=0$. (a) χ vs B ($\chi_{\rm D}$ is the diamagnetic susceptibility for an Al–cluster with $N=10^3$). (b) C vs B for the same cluster.